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PROCESSING-MORPHOLOGY RELATIONSHIPS OF COMPATIBILIZED POLYOLEFIN/POLYAMIDE BLENDS

Part I: The effect of an Ionomer Compatibilizer on Blend Morphology

J.M. Willis and B.D. Favis

Industrial Materials Research Institute

National Research Council Canada

75 De Mortagne Blvd., Boucherville, Que., Canada, J4B 6Y4

ABSTRACT

The morphology of compatibilized polyolefin/polyamide blends was found to be significantly dependent on the concentration of an ionomer compatibilizer (polyethylene-methacrylic acid-isobutyl acrylate terpolymer) in the blend. For a dispersed phase content of 10% by weight, a maximum reduction in phase size was observed when only 0.5% by weight of ionomer was added to the blend. A more significant reduction of the dispersed phase size was observed when the minor phase was nylon, due to interactions which exist between the ionomer and the polyamide. These interactions have been confirmed by Fourier transform infrared spectroscopy. At high concentrations of the ionomer, flocculation of the nylon dispersed phase was observed. In comparison to one-step mixing, blends prepared by two-step or batch mixing were characterized by a smaller dispersed phase when nylon was the matrix, and a larger particle size when nylon was the minor phase. The results observed are explained in terms of a speculative model of the interactions occurring across the nylon-polyolefin interface.

INTRODUCTION

The blending of polymers has become an increasingly important industrial technique. It is an economically viable and versatile way in which new materials can be produced to have a wide range of properties. Several reviews have been written on this subject [1-3].

The majority of polymers are immiscible at the molecular level as given by laws of thermodynamics. However, it is possible to render a blend compatible whereby the resultant blend properties may be synergistic and the morphology of the minor phase in the matrix polymer is finer and more homogeneous. Such compatibilization can be achieved in a variety of ways, the most common of which is the introduction of a third component to the blend which could then promote interactions across the matrix/dispersed phase interface. The interfacial activity of block and graft copolymers has been reviewed by Paul [4]. The compatibilization of immiscible blends by the addition of block copolymers can be compared in some respects to the emulsifying effect of surfactants in oil and water mixtures. The original morphological observations of Molau et al. [5-7] clearly demonstrate the ability of block copolymers to emulsify polymer dispersions in solution and thus inhibit phase separation. As a result of this work and later studies on emulsified blend systems [8, 9], block copolymers are often referred to as oil-in-oil emulsifiers.

The emulsifying effect of block copolymers on polymer blends prepared in solution has been well documented [4]. For practical purposes, this concept has more recently been extended to polymers blended in the melt, by the

addition of graft [10-14], random [15, 16] as well as block copolymers [14, 17-19]. In the latest of a series of papers, Teyssié et al. [20] observed a significant reduction in the dispersed phase size and an increase in the interfacial adhesion as a result of melt blending polyethylene/polystyrene with as little as 2% by weight poly(butadiene-b-styrene) copolymer. The presence of the copolymer also stabilized the dispersion against coalescence. More importantly, Teyssié et al. [21] clearly demonstrated that the copolymer was uniformly adsorbed at the interface between the two polymers.

The object of this study is to determine the emulsifying effect of an ionomer (polyethylene-methacrylic acid-isobutyl acrylate terpolymer), on the morphology of polyolefin/polyamide blends. This compatibilized ternary blend has been proposed as a system for blow moulding applications [22, 23]. For uncompatibilized polyethylene/polyamide blends, Braun and Eisenlohr [24] and Utracki et al. [25] have shown that a distinct two-phase immiscible system is the result of melt blending. However, if maleic anhydride is grafted onto the polyolefin, the interactions across the polyolefin/polyamide interface are significantly enhanced [10, 26, 27]. In this case, an improvement in the impact strength and a reduction in the size of the dispersed polyolefin were observed. Although the compatibilization of polyolefin/polyamide blends by the presence of graft copolymers has been well studied, little work has been done on compatibilized ternary blends such as polyolefin/polyamide/ionomer, despite the fact that some morphology-property studies have been performed on binary blends of polyethylene-based copolymers with polyolefins [28] and polyamides [28-32].

A study of the blend morphology is of central importance because it relates the properties of the blend to the manner in which it was processed. Many studies have dealt with the relationships between morphology, processing, and the physical, rheological and mechanical properties of incompatible blends. For example, it has been reported that polyamide/polyolefin blends may form a laminar, plate-like morphology during blow moulding operations. A lamellar-type morphology has also been observed under certain conditions during sheet extrusion [33]. As shown by Subramanian et al. [34-36] and Kamal et al. [37], blends which are characterized by laminar morphologies may display good barrier properties, depending upon the dispersed polymer. Impact strength is another property which can be improved by blending and which is highly dependent on the size of the dispersed phase. There are several studies which discuss the impact modification of polypropylene [38, 39] and polyamide [11, 12, 26, 40] by blending these polymers with elastomeric materials.

Previous work done in our laboratory has involved the effect of composition [41, 42], viscosity ratio [43] and processing parameters [44] on the dimensions of the minor phase in immiscible binary blends. In this paper, the influence of interfacial modification on morphology is studied. Due to the potential applicability of the compatibilized polyolefin/polyamide/ionomer blend in industry [22, 23], it was of interest to investigate the role of the ionomer compatibilizer as an interfacial or emulsifying agent in these blends in order to determine processing-morphology relationships.

EXPERIMENTAL

Resin Characterization

The polypropylene (PP) resin used in this study was Pro-Fax 6501, obtained from Hercules in powder form. The PP resin was compounded with an antioxidant Irganox 1076, on a twin-screw extruder, and transformed into pellets. The high density polyethylene (PE) sample, 07055C, was obtained from Dow as pellets. The polyamide-6 (PA) sample used in this study was Zytel 211 from Dupont in pellet form. The interfacial agent was Surlyn 9020 from Dupont. This ionomeric resin is a random terpolymer consisting of roughly 80% PE and 20% of a mixture of methacrylic acid, partially neutralized with zinc (approximately 70%), and isobutyl acrylate. The exact composition of this mixture is unknown.

Some properties of these resins are given in Table 1. The torques were determined for each of the pure resins by mixing 60 ml of the resin in the Brabender mixing chamber at 250°C for 5 min at 50 RPM. The storage modulus and dynamic viscosity of each resin were obtained as a function of frequency at 250°C, using a Rheometrics Mechanical Spectrometer (model RMS 605), and are illustrated in Figures 1 and 2, respectively. The instrument was operated in the dynamic mode, with a gap of 1.2 ± 0.2 mm between parallel plates. All experiments were performed under dry nitrogen.

Compounding - One Step

The resins were dry blended and dried under vacuum at 95°C overnight. It may be mentioned that throughout the text, the blend compositions are given in terms of weight fraction.

The mixed resin pellets were then melt blended in the Brabender mixing chamber using the roller blades recommended for high shear applications. A typical blending experiment consisted of the following steps. With the mixing chamber initially set at 250°C and the blades turning at 50 RPM, the resin mixture was fed into the chamber. Once all the resin was added, the blend was allowed to mix for 5 min under a constant flow of dry nitrogen. At the end of 5 min, the melt was rapidly transferred to a mould which was then held in a hydraulic press under 2.5 MPa of pressure until the sample had cooled down to room temperature (approximately 3 min).

Compounding - Two Steps

Master batches of 5% an ionomer compatibilizer in PP, PE and PA were prepared following the same procedure as that outlined above. The moulded sample was then granulated and dry blended with the matrix polymer. The resin mixture was redried overnight under vacuum, and melt blended as above.

Scanning Electron Microscopy

Both freeze fractured preparations and microtomed surfaces were studied on the scanning electron microscope (SEM). In preparation for freeze fracturing, a small rectangular strip of each sample was frozen in liquid nitrogen for 10-15 min.

For the microtomed samples, two separate specimens were cut from the moulded samples to have dimensions of roughly 1 × 1.5 × 0.5 cm. Plane surfaces were prepared on each specimen using a Reichert Jung Supercut 2050 microtome equipped with a diamond knife. Prior to microtoming, the samples were frozen

in liquid nitrogen. While cutting, the surface of the sample was held at approximately -100°C to reduce the degree of surface deformation. It was interesting to note that the surfaces prepared for the uncompatibilized blends displayed very little deformation. In other words, essentially no plowing of the dispersing phase into the surrounding matrix was observed. In contrast, more severe deformation was observed for the compatibilized blends, despite the fact that the microtoming was performed at a temperature as low as -180°C.

The microtomed samples were then treated with a suitable solvent to dissolve the minor phase present at the surface of the specimen. For a nylon minor phase, this procedure consisted of soaking the specimen in m-cresol at room temperature for four days. To dissolve the polyolefin minor phase, the specimens were allowed to sit in decalin at 120°C for 1 hr.

The fracture and microtomed/treated surfaces were then examined under a Jeol 35-CF SEM operating at 10-15 kV.

Image Analysis

The semi-automatic image analyzer used to measure the diameters of the dispersed phase was developed in-house. The operation of this instrument has been described elsewhere [41]. For each sample, six SEM photomicrographs were analyzed, and averages then calculated for the number average diameter, d_n, volume average diameter, d_v, and polydispersity, P_d. A correction factor was applied to the diameters determined from SEM photomicrographs of microtomed surfaces [45]. On average, 600 diameter measurements were taken per blend preparation.

RESULTS AND DISCUSSION

Effect of the Ionomer Compatibilizer on Fracture Morphology

The addition of small quantities of ionomer to blends of PP/PA and PE/PA during one-step compounding resulted in remarkable changes in the morphology of these blends. The fracture surface of a blend of 90% PP/10% PA is Figure 3. The dispersed polymer domains are spherical in shape. There is no evidence of adhesion between the minor phase and the matrix, since the surfaces of the polyamide particles are perfectly clean. When 5% ionomer based on the minor phase was added to 90% PP/10% PA blend, important morphological changes occurred. The fracture surface of this blend is also shown in Figure 3. It can be seen that the dimensions of the dispersed nylon domains have decreased by about three times. It is also evident that the presence of ionomer has improved the adhesion between the two phases, since there appears to be some material coating the surfaces of the dispersed phase.

When the phases of the blend were inverted, some adhesion was also observed on the surfaces of the dispersed PP domains in the 10% PP/90% PA blend containing 5% ionomer whereas no adhesion was observed in the uncompatibilized 10% PP/90% PA blend.

SEM micrographs also indicate a similar improvement in adhesion for the 90% PE/10% PA and 10% PE/90% PA blends, upon compounding with 5% ionomer based on the minor phase.

Dispersed Phase Dimensions

The surfaces prepared by microtoming, followed by minor phase dissolution, are

shown in Figure 4 as observed by SEM. By comparing the morphology of the uncompatibilized polyolefin/polyamide blends with the morphology of the blends containing 5% ionomer by weight, it can immediately be seen that the presence of ionomer in these blends resulted in a decrease in the diameters of the dispersed phase. The results, expressed in Table 2 as reduced particle diameters (d_v (0% ionomer)/d_v (5% ionomer)), show that the extent of particle size reduction was more pronounced when nylon was the dispersed phase. This may be related to the high affinity which the ionomer has for polyamide, and will be discussed in a subsequent section of this paper.

Effect of Ionomer Concentration on Phase Size

The reduction in the dimensions of the dispersed phase was studied as a function of the quantity of ionomer added to the blend during compounding. Figure 5 illustrates the dependence of the number average and volume average diameters upon the ionomer content for blends of 90% PP/10% PA and 10% PP/90% PA. Similar curves were obtained for 90% PE/10% PA and 10% PE/90% PA blends, as shown in Figure 6. In all cases, it appears that approximately 5% ionomer based on the minor phase is sufficient to produce a maximum reduction of the dispersed phase size. No further decrease in phase size is achieved by adding more ionomer, as seen from the plateau regions of the plots shown in Figures 5 and 6. Teyssié et al. [20] also observed an equilibration in the phase size, when increasing amounts of block copolymer were added to polyethylene/polystyrene blends. In their study, the most significant effect of size reduction occurred from 0.5% to 1.0% by weight of copolymer (based on the blend).

It is well known from emulsion studies that the dispersed particle size diminishes when surfactant is added to the mixture [46]. This effect is due to the ability of the surfactant to reduce the interfacial tension between the dispersed phase and the matrix. The quantity of surfactant required to fully cover an interface is related to such variables as the degree and type of mixing, the affinity of the surfactant for the dispersed phase, the size of the dispersed phase, the rate of adsorption and orientation of the surfactant at the interface, and the extent to which the interface is stabilized against flocculation and subsequent coalescence. The deformation of the droplet is also very important. In essence, a droplet will deform until the total force acting on the particle becomes equal to the surface tension. In a theoretical treatment of particle deformation, Taylor [47, 48] developed a variable, We, defined as

$$We = \eta_m d_n \dot{\gamma} / 2\gamma_{12} \qquad (1)$$

where η_m is the viscosity of the matrix or continuous phase, d_n is the number average diameter of the dispersed phase, $\dot{\gamma}$ is the shear rate or velocity gradient during mixing, and γ_{12} is the interfacial tension. It can be envisaged from Equation (1) that there is a critical value of We below which particle deformation will no longer occur, and as a result, a critical particle size. At this point, the emulsion system has achieved the maximum interfacial area possible. Therefore, there must be a maximum quantity of surfactant which can saturate the interface. Addition of surfactant beyond this concentration will not give rise to further particle size reduction.

This has recently been demonstrated by Djakovic et al. [49] for oil/water emulsions.

For the compatibilized polyolefin/polyamide blends, it appears that the equilibrium concentration of ionomer, beyond which significant particle size reduction no longer occurs, is 5% by weight of dispersed phase. In fact, this corresponds to only 0.5% based on the weight of the blend.

An equilibrium phase size of 0.5 ± 0.1 μm was obtained for the 90% PP/10% PA, 90% PE/10% PA, and 10% PE/90% PA blends as a result of the emulsifying effect of the ionomer. In contrast, the equilibrium phase size observed for 10% PP/90% PA blend was twice as large, 1.1 μm. It is possible that the ionomer has less affinity for the dispersed polypropylene, in comparison to dispersed polyethylene and nylon. Thus, the interfacial tension of the dispersed PP blend would not be reduced to the same extent as for the dispersed PE and PA blends, and a larger equilibrium phase size would result.

Particle Size Distribution

The dimensions of the minor phase not only decrease but also attain a more uniform distribution in blends which contain ionomer in comparison to blends without ionomer. This is shown in Figures 7 and 8 for PP/PA and PE/PA blends, respectively. The percent cumulative (linear) distributions are very broad for the blends which do not contain ionomer. These distributions become significantly narrower when ionomer is added. In fact, the reduction in the dispersity of the particle diameter can be as much as 30%, when 5% ionomer is added to the blends during compounding. The particle size distributions

become only slightly narrower when larger quantities of ionomer are added. These results can also be correlated to emulsion studies, where the addition of surfactant has been shown to narrow the particle size distribution until a certain concentration of emulsifier has been reached [49].

Blend Viscosities

The variation in the torque of the blend as a function of the ionomer content is shown in Figure 9. For all four blends studied, the values measured for the torque increased as the quantity of ionomer in the blend increased. Since the torque gives an indication of the complex viscosity of the material within the mixing chamber (at a given shear rate), the trends shown in Figure 9 indicate that the blends become more viscous when ionomer is added during compounding. It is well known that an incompatible blend, characterized by a sharp interface and essentially no interaction between the two phases, frequently exhibits interlayer slip. This gives rise to a reduction in the viscosity of the blend (negative deviation from additivity) [50, 51]. The fact that the torques and thus the viscosities have increased indicates that there is less slippage at the interface as a result of the addition of ionomer [51]. This suggests that there are strong interactions at the interface between the polymers.

Specific Interactions

Evidence has been presented in this study to suggest that there are interactions which occur preferentially between the polyamide and the ionomer. These interactions are apparently occurring to a much smaller extent

between the ionomer and the polyolefins. This is particularly true for polypropylene, since the equilibrium particle size was shown in Figure 5 to be twice as large for the PP dispersed phase blend in comparison to the other blends.

It has recently been shown using Fourier transform infrared spectroscopy (FTIR) that there is a significant degree of hydrogen bonding between the amine groups of nylon and the carbonyl groups of the ionomer, particularly at low concentrations of ionomer[52]. This is reproduced in Figure 10. Binary blends of the polyolefins with the ionomer do not display such an effect. In contrast, the intermolecular hydrogen bonding of the ionomer is disrupted by the presence of the polyolefins. These results allude to stronger interactions between the ionomer and the polyamide than between the ionomer and the polyolefins and support the morphological observations reported in Figures 3 and 4. Such strong intermolecular interactions have been shown to play an important role in the compatibilization of polymer blends [53].

In a study of the binary blends of polyamide-6 and ethylene-methacrylic acid copolymers, McKnight et al. [30] suggested that graft copolymers were formed between the two phases as a result of amidation reactions taking place during melt mixing. Similar results were obtained by Chuang and Han [29, 31-32] for binary blends of nylon-6 with ethylene-based multifunctional polymers. It is interesting that no evidence of such graft reactions was observed for binary blends of the ionomer and nylon [52]. This is probably due to the fact that more than 70% of methacrylic acid groups of the ionomer are neutralized by zinc. The salt form of the acid would thus inhibit the amidation reaction.

Flocculation of Dispersed Phase at High Ionomer Concentrations

An interesting phenomenon was observed at the higher ionomer concentrations, when the dispersed phase was nylon. For blends of 90% PP/10% PA and 90% PE/10% PA containing 15% and 30% ionomer (based on the minor phase), aggregation of the dispersed phase was observed. This is shown in Figure 11, for both fracture and microtomed/treated surfaces. It appears from these observations that once the particle size has stabilized, the excess ionomer macromolecules tend to concentrate together at the matrix/nylon interface and produce aggregates of dispersed nylon during mixing.

In order to confirm the composition of the material existing between the flocculated dispersed phase, X-ray spectrometric analyses (EDX) were carried out on the SEM. As shown in Figure 12, a high concentration of oxygen was observed in the interparticle spaces within the floc, in comparison to the matrix phase. Since the nylon had been selectively removed prior to the SEM analysis, this observation identified the interparticle material as the ionomer.

From emulsion studies, it is known that flocculation arises due to strong interparticle interactions [54]. For ionomers such as Surlyn, the formation of ion clusters is a frequently observed but poorly understood phenomenon [55, 56]. It is possible that the interactions which promote the formation of ion clusters are also responsible for the flocculation observed for the 90% PP/10% PA and 90% PE/10% PA blends containing large quantities of an ionomer compatibilizer.

Since generating a uniform dispersion is one of the functions of blending, these observations may be important, particularly with respect to applications dealing with high concentrations of ionomer. The formation of aggregates and the stabilization of the particle diameter at an ionomer concentration of 5% (by weight of dispersed phase) indicate that the addition of excess quantities of ionomer is not necessary and may perhaps have a detrimental effect on blend properties.

Mode of Addition of the Interfacial Agent

In a recent morphological study of nylon/rubber blends, Cimmino et al. [11] observed a reduction in particle size when the blends were prepared in two steps in comparison to one-step mixing. Two-step mixing was performed by first preparing a master batch of the polymer destined to be the minor phase containing the interfacial agent. This master batch was then compounded with the matrix polymer. By concentrating the interfacial agent directly in the polymer which would subsequently be the minor phase, it is possible that the interactions which occur across the interface may proceed more readily. It was of interest to determine whether compatibilized polyolefin/polyamide blends prepared in this manner would have a finer dispersion, or in other words, a smaller dispersed phase size, and more uniform morphology. The effect of two-step mixing on blend morphology was studied for both nylon matrix and polyolefin matrix blends. The fracture surfaces observed for these blends are shown in Figure 13. The dimensions of the dispersed phase, measured from microtomed/treated surfaces, are given in Table 3.

For the nylon matrix blends, containing a polyolefin dispersed phase

preblended with 5% ionomer (0.5% by weight of blend), a 15% decrease in the particle size was observed for two-step mixing in comparison to one-step mixing. This suggests that the ionomer is somewhat more concentrated at the interface by blending in two steps.

When the phases are inverted (i.e., nylon dispersed phase), two-step mixing resulted in significantly larger phase dimensions. This is contrary to what was originally expected. For these blends, preblending the nylon dispersed phase with the ionomer increased the particle size by an average of 68% relative to one-step mixing.

These observations may be interpreted in terms of the following speculative mechanism. It has previously been mentioned that the ionomer has a high affinity for nylon. By preblending the ionomer with the nylon dispersed phase, it is possible that during melt mixing, this affinity would ensure a random distribution of the ionomer within the nylon dispersed phase rather than a high concentration of ionomer at the nylon/polyolefin interface. This is shown schematically in Figure 14. Since the interactions across the interface have diminished, two-step mixing in this situation is disadvantageous. For the nylon matrix blends, where the ionomer is preblended with the polyolefin dispersed phase, the high affinity of the ionomer for the polyamide may result in a preferential concentration of ionomer at the nylon/polyolefin interface during mixing. A speculative representation of this effect is also shown in Figure 14. Therefore, in this situation, it is advantageous to undertake two-step mixing.

It may be mentioned that there is a negligible change in the torque ratio when comparing the blends prepared in one step to those prepared in two steps. In other words, at the concentrations of ionomer used in this study, the presence of ionomer has little influence on the viscosity of the components of the blend. This was determined recently in a study of the binary blends of the ionomer with PP, PE and PA [52]. Therefore, this suggests that the influence of the mode of mixing on the size of the dispersed phase is related to interfacial phenomenon, as described in Figure 14, rather than rheological effects.

CONCLUDING REMARKS

From SEM micrographs of fracture surfaces, the addition of the ionomer to polyolefin/polyamide blends was found to increase the adhesion between the matrix and the dispersed phase. The compatibilized blends displayed a much finer dispersion of the minor phase in the matrix polymer. The emulsifying ability of the ionomer also caused a reduction in the polydispersity of the particle size. It was of interest that as little as approximately 0.5% ionomer by weight of the blend was sufficient to produce a maximum reduction of the dispersed phase size. Larger quantities of ionomer yielded aggregates of the dispersed polyamide, possibly due to the clustering phenomenon of ionomers. The hydrogen bonding interactions occurring between the ionomer and the polyamide, as observed from FTIR, resulted in a greater reduction in the phase size of dispersed nylon in comparison to dispersed polyolefin. The mode of addition of the interfacial agent was found to be an important parameter determining the resultant blend morphology. A speculative description of the interactions which may exist across the matrix/dispersed phase interface was

developed to interpret the morphological observations made on these blends.

Future work will involve an analysis of the effect of the ionomer on the morphology of polyamide/polyolefin blends as a function of both the blend composition and the viscosity of the components.

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Table 1 Characterization of the Surlyn, polyethylene, polypropylene and nylon-6 resins

Resin	Melt Index (g/10 min)	Density @ 250°C ($g\ ml^{-1}$)	Torque @ 250°C after 5 min (N m)	Dynamic Viscosity @ 250°C 100 rad/s $\times 10^{-2}$ (Pa·s)	Storage Modulus @ 250°C 100 rad/s $\times 10^{-4}$ (Pa)
Surlyn	1.0	0.74	14.3	5.0	4.6
PE	7.0	0.74	5.9	3.0	1.5
PP	4.0	0.75	8.6	3.2	3.6
PA	-	0.96	8.6	6.2	2.6

Table 2 **Reduction in phase size achieved through the compatibilization of polyolefin/polyamide blends by the addition of 5% ionomer (by weight based on the minor phase).**

Matrix 90%	Dispersed Phase 10%	$\frac{d_v\ (0\%)^*}{d_v\ (5\%)}$
PA	PP	2.3
	PE	2.8
PP	PA	4.2
PE		4.1

* Reduced diameter ≡ Ratio of diameter of uncompatibilized blend to that of blend containing 5% ionomer.

Table 3 Comparison of the number average diameter measured for polyolefin/ polyamide blends prepared in two steps with those prepared in one step.

Sample Composition		Number Average Diameter (μm)		
Matrix Polymer	Dispersed Phase Polymer	Without ionomer	With 0.5% ionomer (by weight of blend)	
			One step mixing	Two step mixing
PA	PP	3.12	1.53	1.29
	PE	1.78	0.55	0.47
PP	PA	2.69	0.81	1.11
PE		3.12	0.73	1.49

FIGURE CAPTIONS

1. Storage modulus, G', as a function of frequency, measured for the pure resins at 250°C.

2. Dynamic viscosity, η', as a function of frequency, measured for the pure resins at 250°C.

3. Fracture surfaces of a 90% polypropylene/10% polyamide blend:
   a) without ionomer
   b) with 5% ionomer by weight based on the minor phase.

4. Surfaces prepared by microtoming, followed by minor phase dissolution. Comparison between the uncompatibilized blends and blends containing 5% ionomer by weight based on the minor phase: a) 90% PP/10% PA, b) 10% PP/90% PA, c) 90% PE/10% PA and d) 10% PE/90% PA.

5. The dependence of the number and volume average diameters measured as a function of the ionomer concentration for: a) 90% PP/10% PA and b) 10% PP/90% PA.

6. The dependence of the number and volume average diameters measured as a function of the ionomer concentration for: a) 90% PE/10% PA and b) 10% PE/90% PA.

7. Cumulative (linear) distributions of the particle size measured for: a) 90% PP/10% PA and b) 10% PP/90% PA blends containing 0%, 5% and 30% ionomer by weight based on the minor phase.

8. Cumulative (linear) distributions of the particle size measured for: a) 90% PE/10% PA and b) 10% PE/90% PA blends containing 0%, 5% and 30% ionomer by weight based on the minor phase.

9. Variation in the torque measured for polyolefin/polyamide blends studied, as a function of the ionomer concentration.

10. The dependence of the ratio of the area of the absorption band at 1699 cm^{-1} (representing hydrogen bonded carbonyl groups) to the band at 1735 cm^{-1} (representing non-hydrogen bonded carbonyl groups) upon the weight percent of ionomer in binary blends of ionomer with nylon, polyethylene and polypropylene, as determined by transmission FTIR [52].

11. Fracture (A) and microtomed/treated (B) surfaces of a 90% PP/10% PA blend containing 30% ionomer by weight of nylon, illustrating the aggregate formation.

12. Energy spectra obtained for the X-ray spectrometric analysis of 90% PE/10%PA containing 30% ionomer by weight based on the dispersed phase, illustrating the difference in oxygen content between the matrix and the interparticle regions within the aggregates. The sample analyzed was microtomed followed by minor phase dissolution and carbon coated prior to EDX analysis on the SEM.

13. Fracture surfaces of the compatibilized blends prepared in two steps:

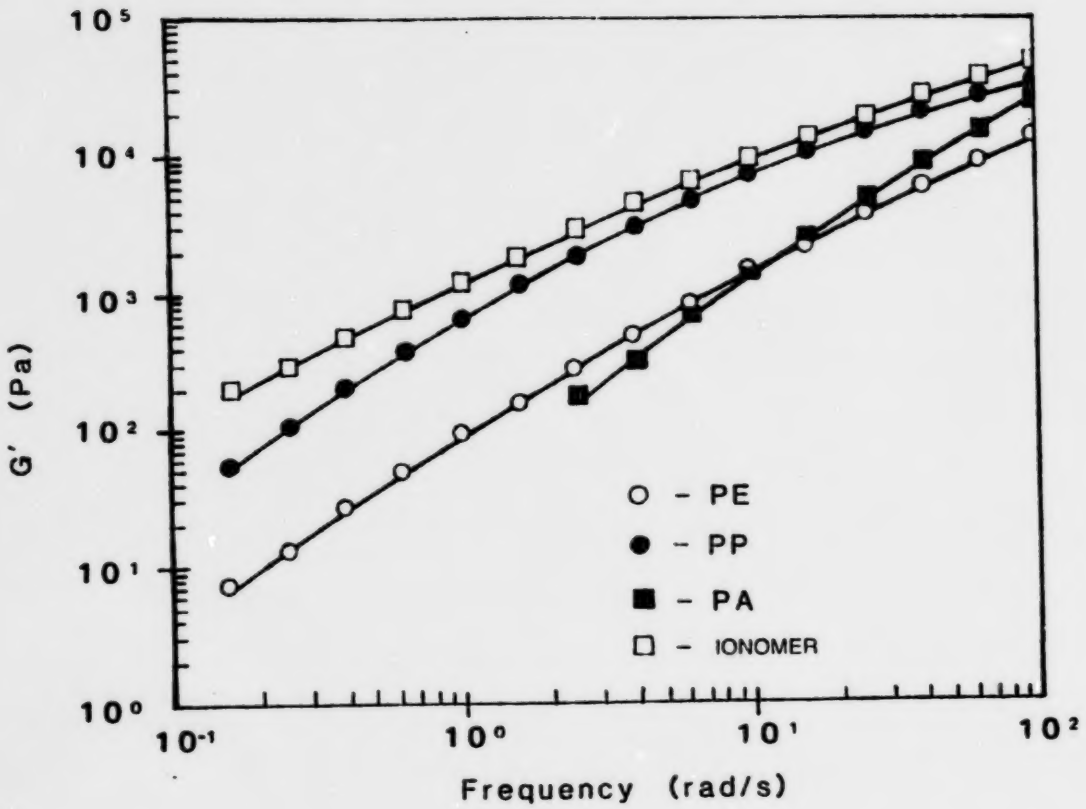
    a) 90% PP/10% (PA preblended with 5% ionomer)

    b) 90% PA/10% (PP preblended with 5% ionomer)

    c) 90% PE/10% (PA preblended with 5% ionomer)

    d) 90% PA/10% (PE preblended with 5% ionomer).

14. Speculative model of the interactions between the ionomer and the matrix and dispersed phase polymers, during one and two step mixing. The stars represent the ionomer, and the arrows represent figuratively the interactions across the polyolefin/polyamide interface.



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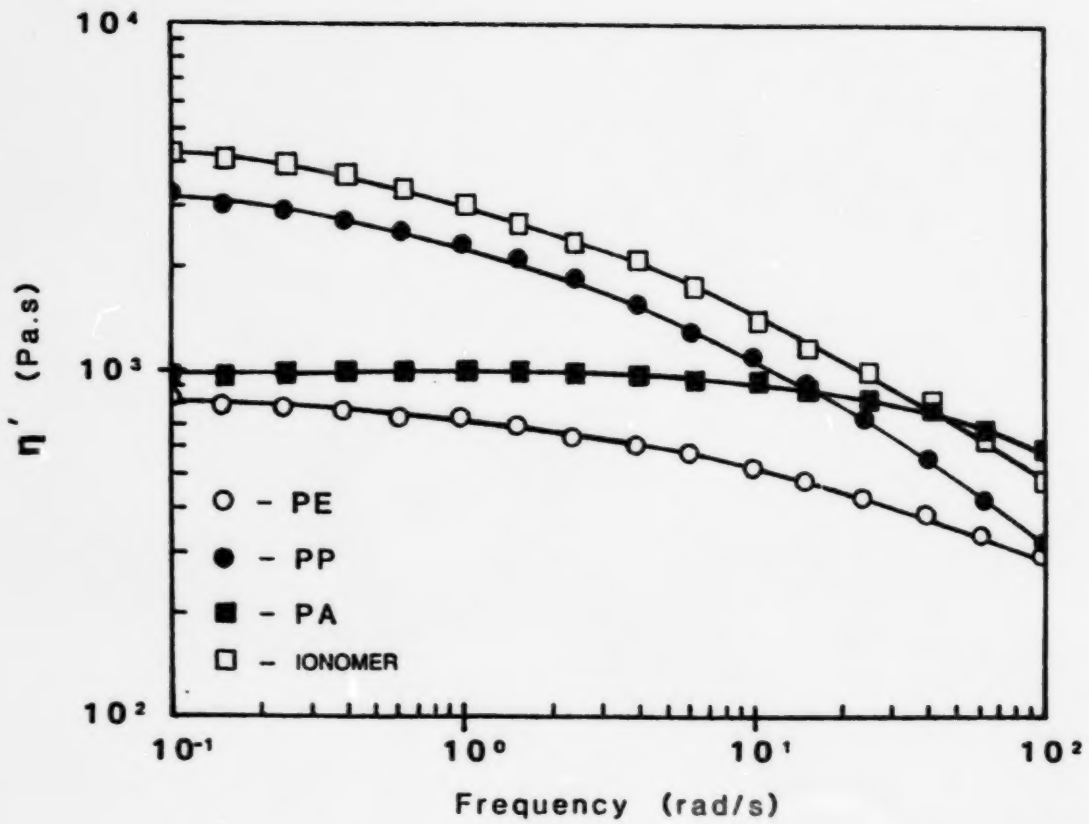

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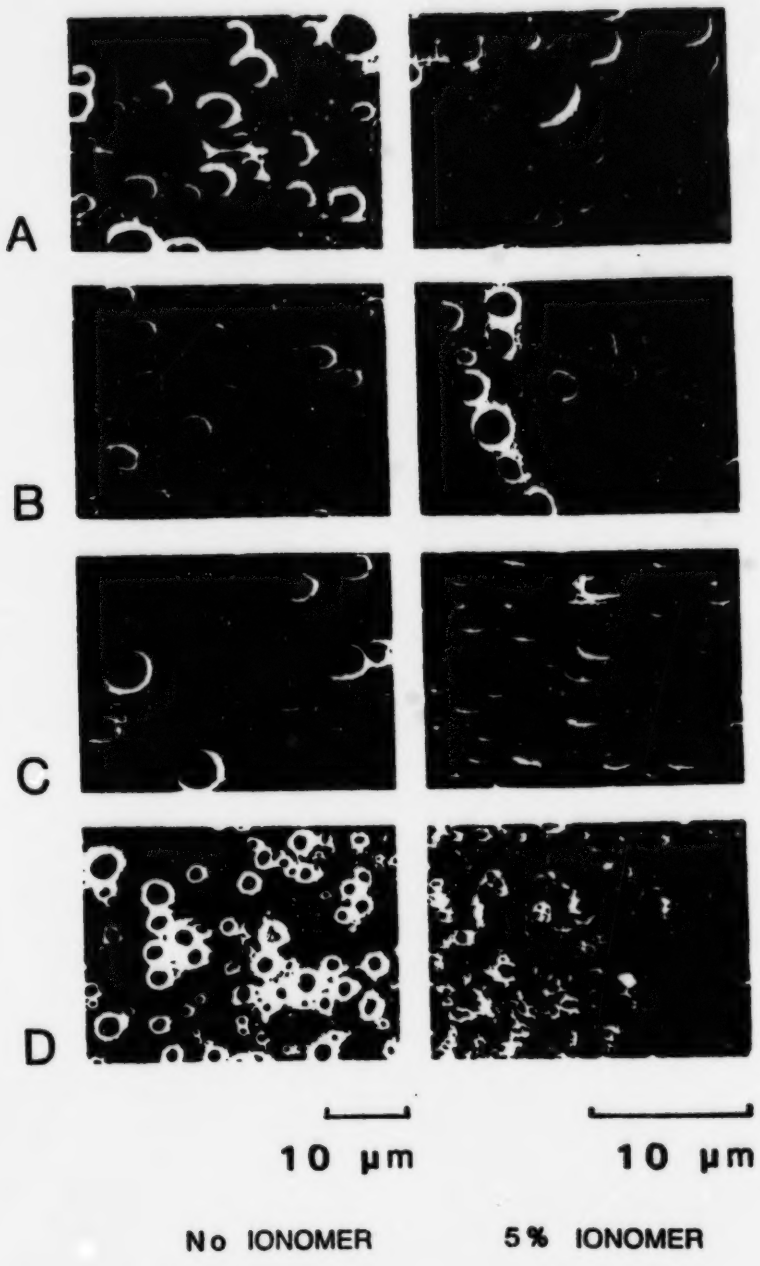

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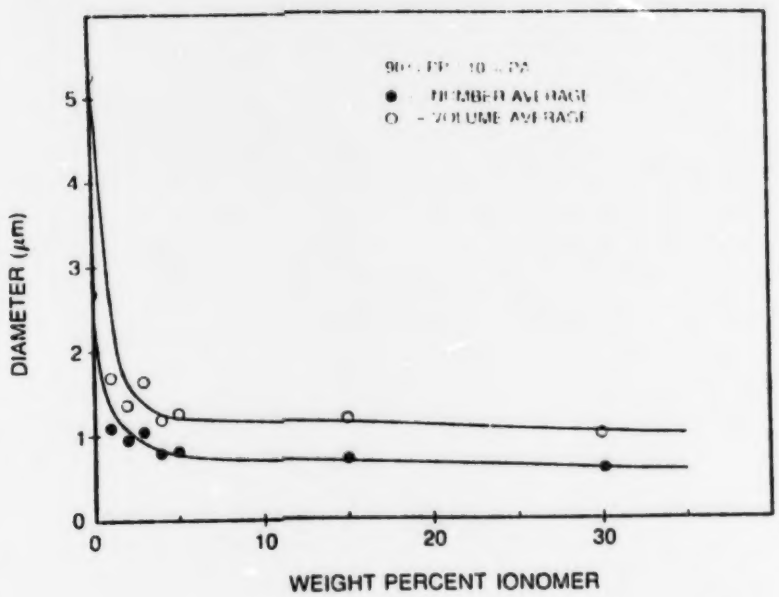

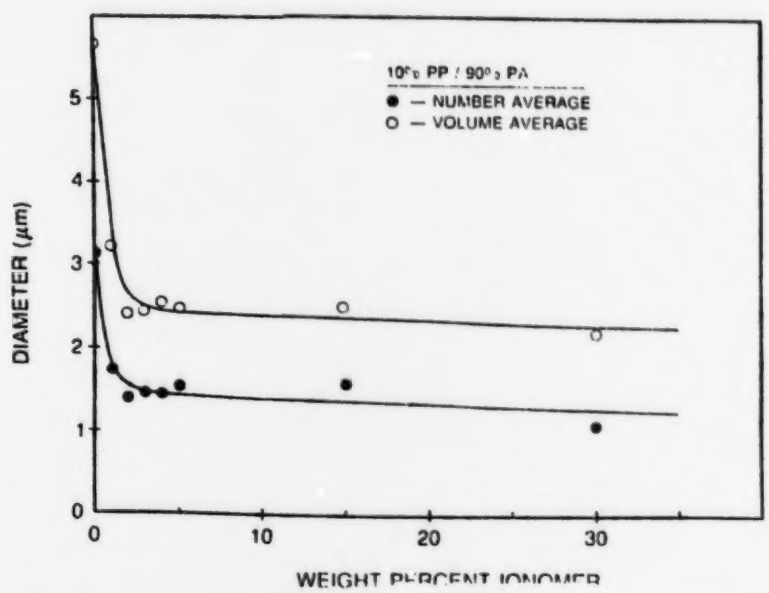

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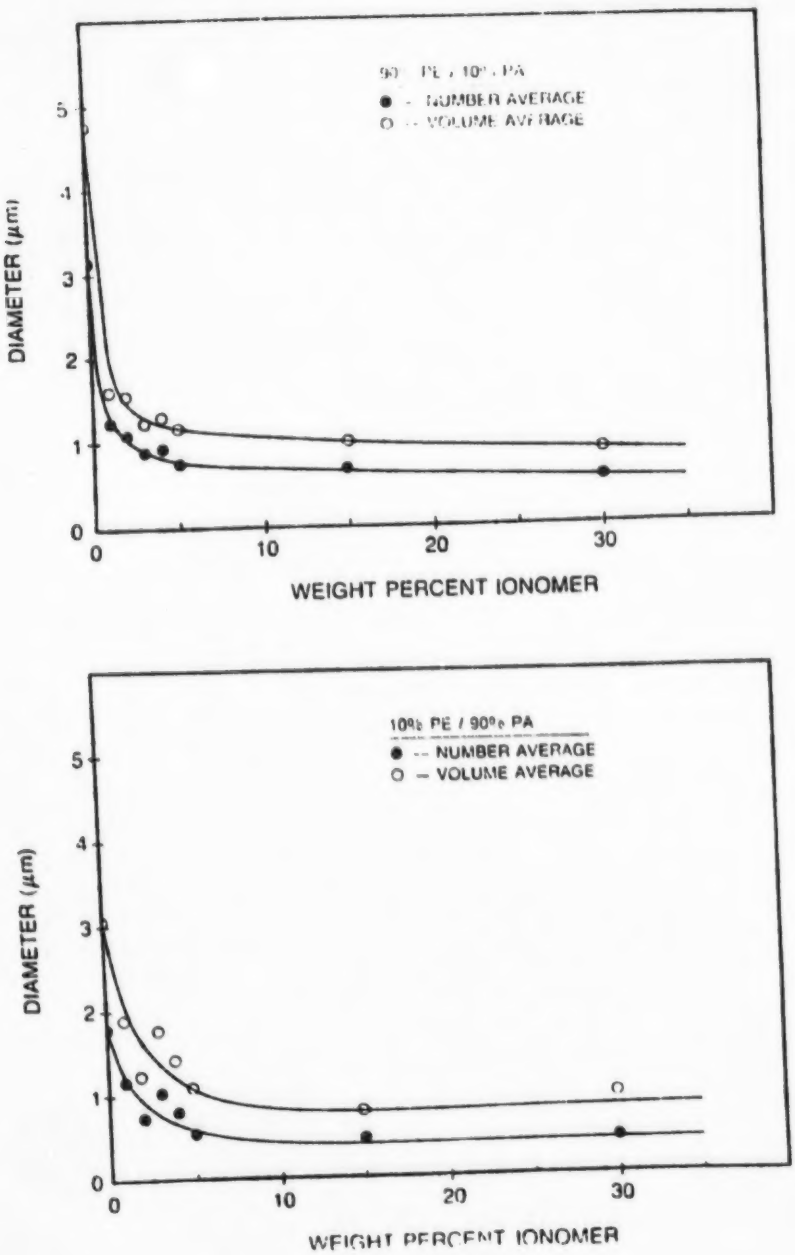

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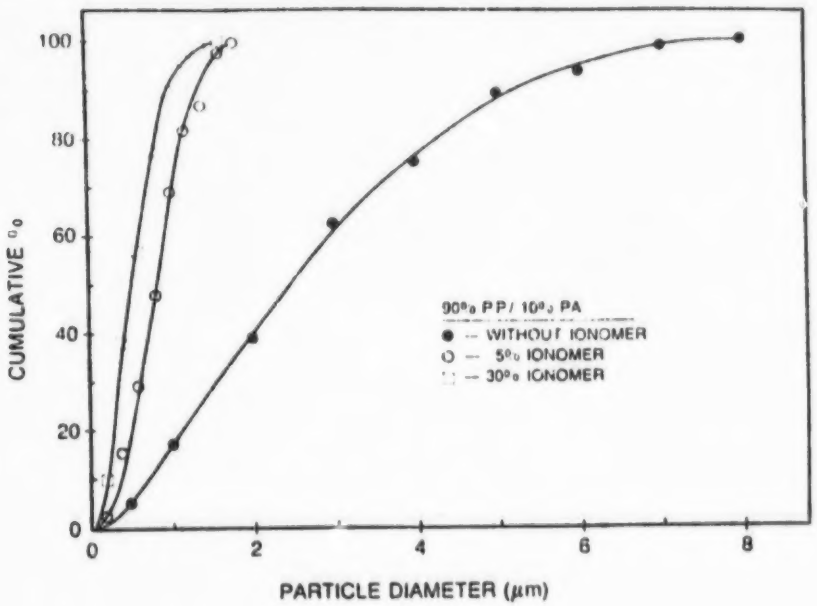

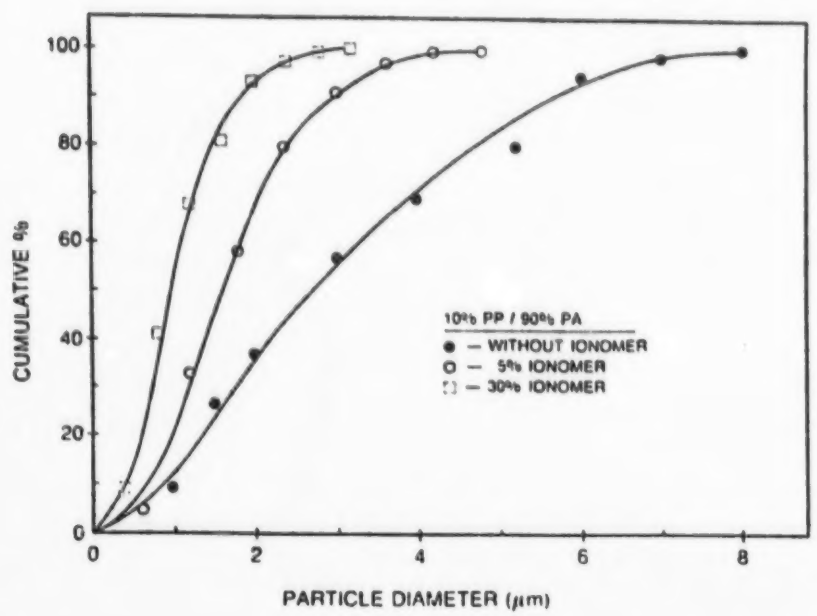

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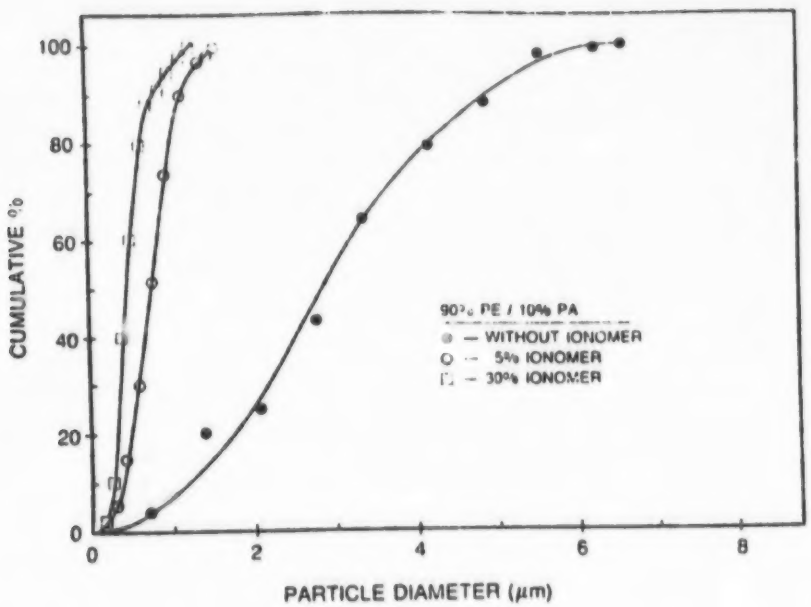

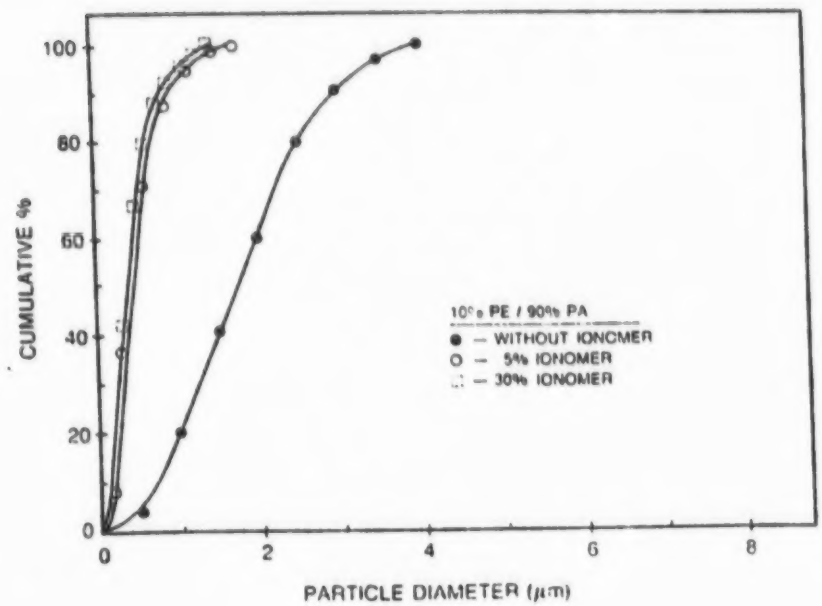

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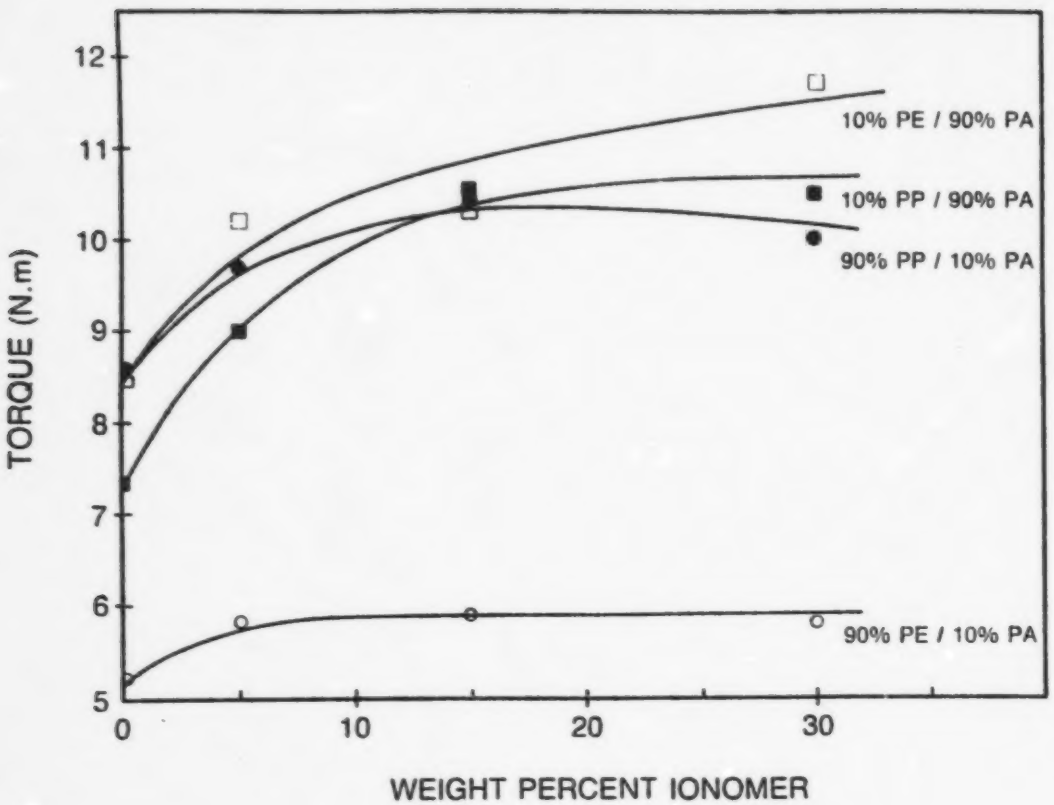

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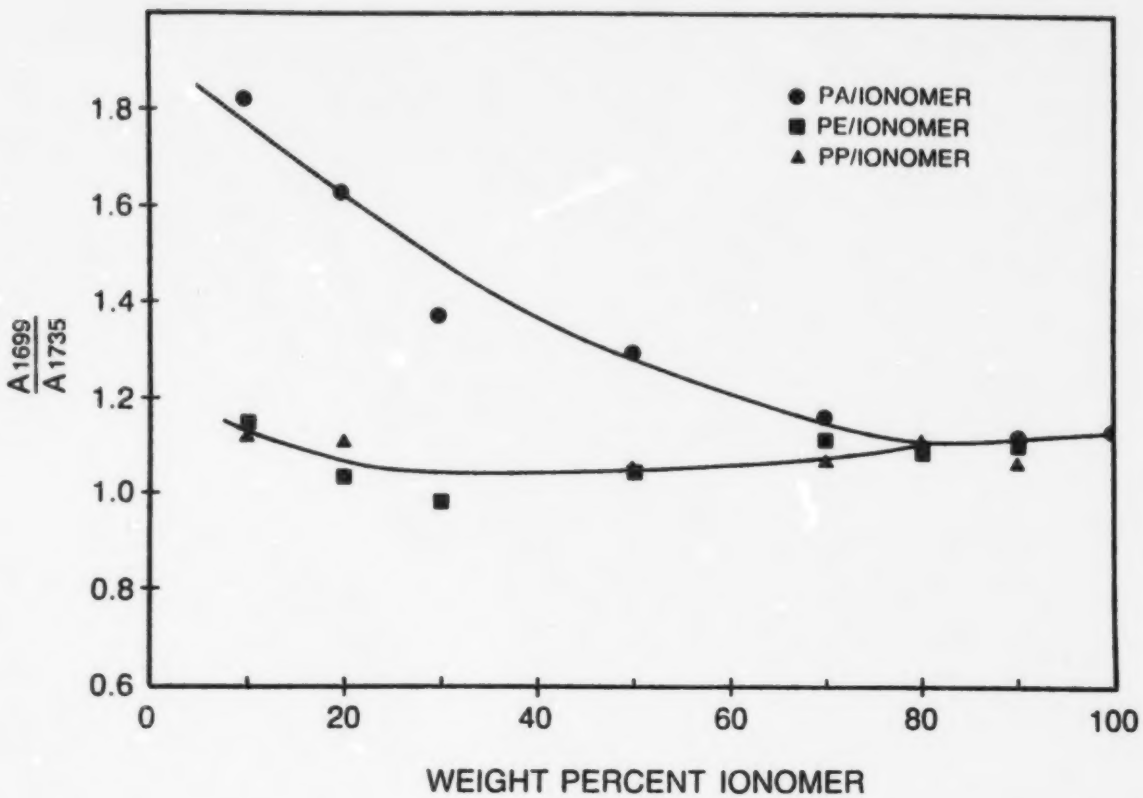

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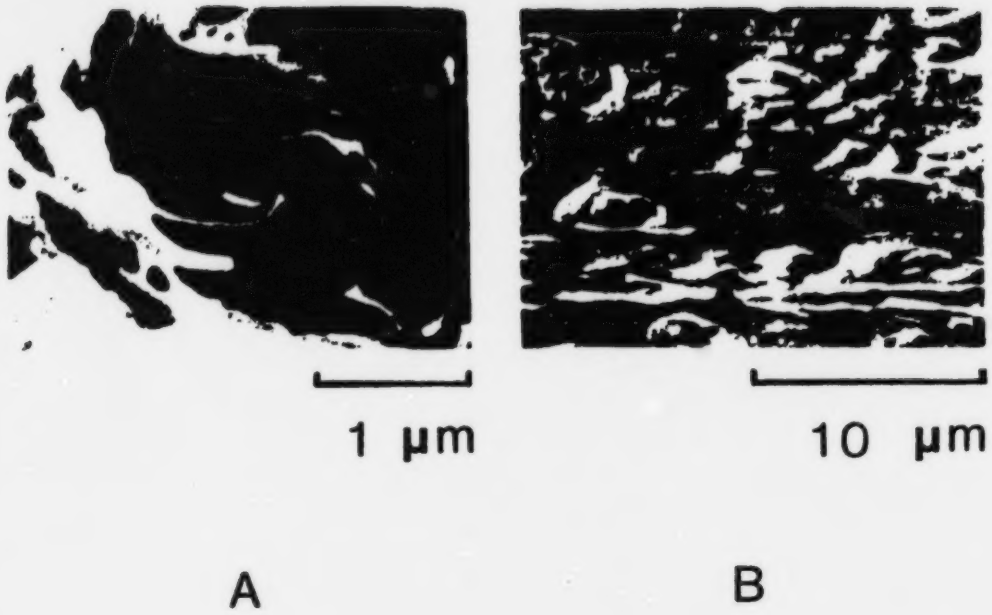

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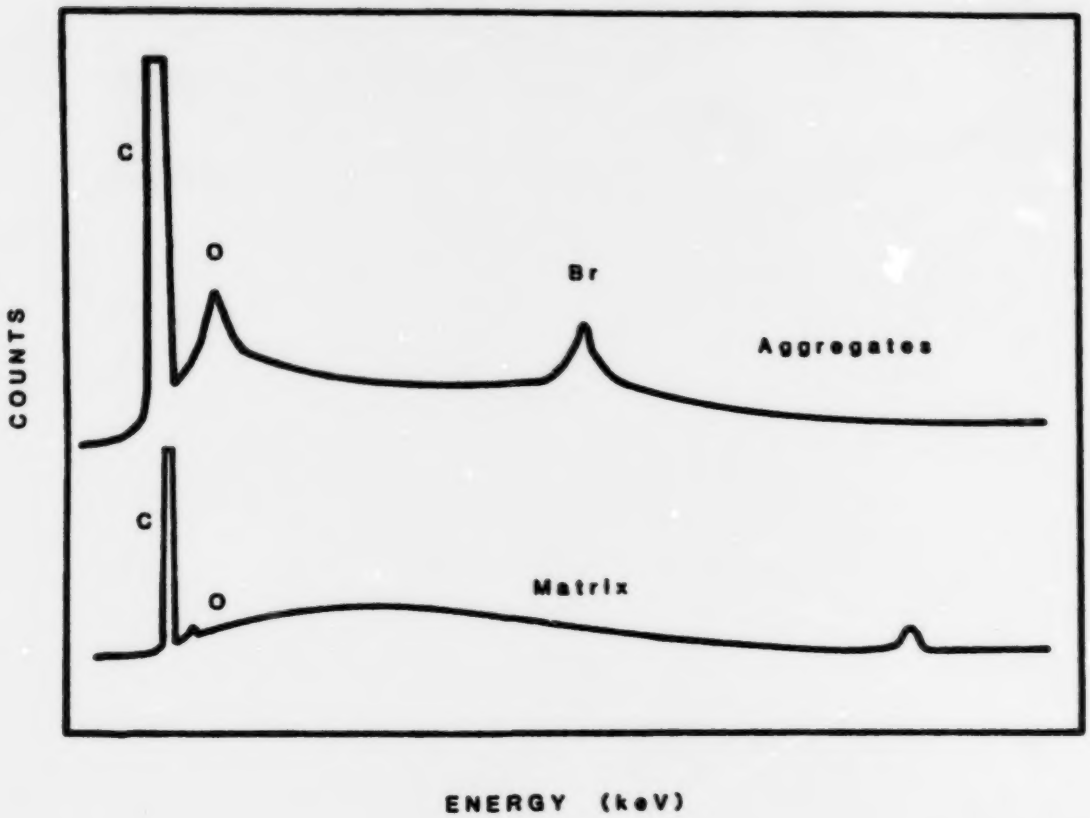

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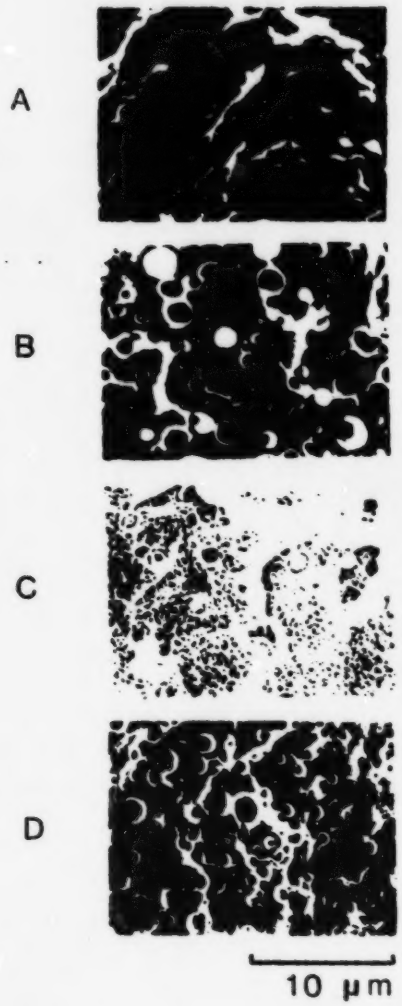

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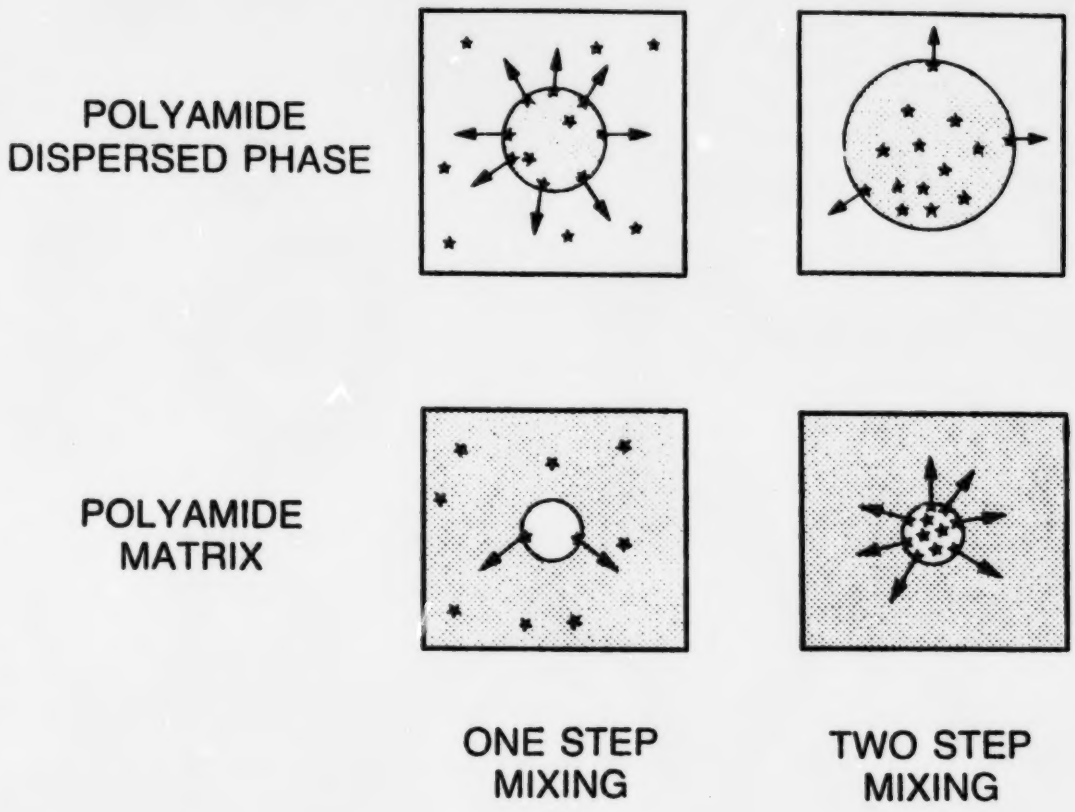

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Discussion: J.M. WILLIS

A.P. PLOCHOCKI: The conceptual part of your work was the use of an ionomer as a coupling/compatibilizing ingredient. The original Dupont patent addressed improvement of barrier properties due to introduced morphological changes. In your micrographs the domains were spherical, not lamellar as the patent would stipulate. My feeling is that this was due to improper elasticity ratio.

J.M. WILLIS: The main objective of this study was the effect of an interfacial agent on blend morphology. The type of mixing we performed was not similar to processing by a blow molding application. We prepared the blend in order to have spherical inclusions. During blow molding these domains would be biaxially stretched into lamellas. However, in our work, we did not perform any blow molding. The elasticity ratios in the systems we studied ranged from 0.6 to 1.7.

M. XANTHOS: Do you know if there is a chemical reaction between PA and ionomer?

J.M. WILLIS: According to FTIR there is hydrogen bonding. There is no evidence of chemical reaction. In addition, the ionomer used in this work was almost completely neutralized, and as such would inhibit any reaction.

P. LEPOUTRE: I have a problem reaching a stable temperature in a 250 ml chamber. It takes 15 minutes to reach the operating temperature. Do you

know how to shorten this period.

J.M. WILLIS: Preheat the rotors and blends.

G.W. LOHFINK: Did you check the morphology before compression molding?

J.M. WILLIS: The micrographs in this study were all obtained after compression molding. The molding was done very rapidly with small deformation.

B.D. FAVIS: In response to the same question we have done a series of experiments on various blends to evaluate the influence of our preparation technique on morphology. We have taken the material from the mixing chamber where one part was quenched and the other part compression molded. We observed the same morphology in both materials.